Ambient atmosphere effect on dry laser cleaning efficiencies for sub-micron particles

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The effects of viscous air drag and Brownian particle motion on dry laser cleaning efficiencies are discussed for $\sim 10^{-2}$-1 μm spherical particle contaminants on Si substrates for initial particle velocities of ~ 10-10^3 m/s and characteristic cleaning times of $\sim 10^{-2}$-10^2 seconds.

The efficiency of dry laser cleaning (DLC) of critical surfaces under ambient conditions is a crucial problem for removal of contaminants smaller than 0.1 micron. Such particulate contaminants are the "killer defects" for ICs, masks and high density memory devices.[1] Complete (~100%) removal of monodisperse sub-micron particles from a smooth substrate seems to be intuitively evident for laser fluences just slightly above the corresponding well-defined cleaning threshold, F_{DLC}, and has indeed been observed in vacuum.[2] In reality, recontamination of a critical surface by removed particulates occurs and DLC efficiency dependence on laser fluence at ambient conditions exhibits typically a non-linear S-like character (Fig.1).[3] DLC efficiency increases asymptotically with increasing laser fluence with less-steep slopes for smaller particles[2-4] (cf. Fig.1) to a percentage limited by the damage threshold fluence for the substrate of interest. Higher cleaning efficiencies may be also achieved with multi-shot repetitive laser irradiation[4] and auxiliary removal tools such as a gas jet,[5] vacuum removal or thermophoresis[6]. However, although DLC technological goals require solution of the contaminant redeposition problem,[4,6] a comprehensive analysis of this problem has not yet been performed.[7] In this paper we propose a model describing recontamination of a critical surface by removed particles via their viscous air drag deceleration and Brownian motion to the surface for a wide range of particle sizes, lift-off velocities and characteristic cleaning times.

The model is based on the intuitive assumption of ~100% DLC of monodisperse, spherical particles from a smooth, vertically oriented substrate (e.g., a Si wafer) in vacuum due to macroscopic thermal "trampoline" or "hopping" effects.[1,8] These sub-micron particles of a radius R and a mass $M=(4\pi/3)\rho R^3$ are assumed to have an arbitrary mass density, ρ, of $\sim 10^3$ kg/m^3 and a surface density, N_0. The particles being cleaned from an ~1 cm-wide surface area illuminated by a homogeneous laser beam leave the surface van der Waals potential well with initial lift-off ve-

locity ΔV_0, i.e., the velocity in excess of the critical velocity, V_{crit}, necessary to overcome the particle/substrate adhesion. Due to the viscous air drag force removed particles are decelerated and their assumed initial δ-function spatial particle distribution is broadened. In this work the broadening effect was neglected relative to the deceleration, assuming that the δ-function character of the spatial particle distribution is maintained up to a certain maximum lift-off distance, X_0, from the substrate surface. As the substrate serves as a "trap" for removed particles with a presumed ~100%-sticking probability, one can consider one-dimensional Brownian motion (diffusion) of the removed particles to the substrate,[9] i.e., diffusion-limited adsorption. A redeposition or exposure time, T_{exp}, for the substrate is determined primarily by the time between cleaning pulses and the scanning velocity of the substrate relative to the laser spot, i.e., the time available for removed particles to diffuse back onto the substrate. For simplicity, we assume that particles that are removed remain suspended while the substrate is moved, ignoring the substrate/ambient shear force. Typical DLC cycle times, T_{exp}, are ~10^{-2} s, when a contaminated area is scanned during DLC with a moderate velocity of ~1 m/s, to ~10^{2} s under stationary conditions.

Disregarding in a first approximation auxiliary transport of laser-removed particles by suction, gas jet or thermophoresis, one can obtain the spatial particle density $n(x,t)$[9] at a distance x from the lift-off point, X_0, which is defined as the x-coordinate origin, x=0,

$$n(x,t) = \frac{N_0}{\sqrt{\pi D(R)t}} \exp\left(\frac{-x^2}{4D(R)t}\right), \tag{1}$$

which is the solution of the one-dimensional Fokker-Planck equation without a convection term and with the following initial and boundary conditions in the absence of the substrate

$$\frac{\partial n(x,t)}{\partial t} = -\frac{\partial J(x,t)}{\partial x} = D(R)\frac{\partial^2 n(x,t)}{\partial x^2}; \quad n(0,0) = N_0\delta(x); \quad n(x<0,t) = n(\infty,t) = 0, \tag{2}$$

where $D(R)=kT/3\pi\eta_{air}R$ is the particle diffusion coefficient,[9] k is the Boltzmann constant, $T\approx300$ K and $\eta_{air}\approx\eta_{N2}(300K, 1atm)\approx2\times10^{-5}$ Pa·s[10] are the air temperature and viscosity under ambient conditions, respectively. Particle redeposition on the substrate may be taken in account by integrating over time the particle flux, $J(x,t)$, at the position of the substrate surface, $x=X_0$, or, at a particular time, integrating the spatial distribution of particles diffused "behind" the substrate surface

$$N_S(X_0,t)=\int_0^t J(X_0,t')dt'=\int_{\infty}^{X_0} n(x,t)dx=N_0\,\mathrm{erfc}\left(\frac{X_0}{2\sqrt{D(R)t}}\right). \quad (3)$$

Then a cleaning efficiency, E, for the given T_{exp} is

$$E(X_0,R,T_{exp})=\frac{N_0-N_S(X_0,T_{exp})}{N_0}=\mathrm{erf}\left(\frac{X_0}{2\sqrt{D(R)T_{exp}}}\right). \quad (4)$$

To evaluate characteristic lift-off distances, deceleration of particles due to the viscous air drag force was considered at low Reynolds number, $Re=\rho_{air}R\Delta V_0/\eta_{air}\sim1$, i.e., for Stokes flow[11]

$$\Delta V(t)=\Delta V_0\exp\left(\frac{-t}{\tau_{air}(R)}\right);\quad x(t)=\Delta V_0\tau_{air}(R)\exp\left(\frac{-t}{\tau_{air}(R)}\right), \quad (5)$$

where $\tau_{air}(R)=2\rho R^2/9\eta_{air}$ is the characteristic velocity relaxation time. For particle sizes of 10^{-2}-1 μm Stokes flow occurs for ambient conditions (air mass density $\rho_{air}\approx1$ kg/m^3, the air viscosity $\eta_{air}\approx2\times10^{-5}$ Pa·s) and maximum initial particle velocities of ~10 or 10^3 m/s attainable for metals or semiconductors using ns or ps/fs laser pulses,[12] respectively. Corresponding lift-off times $t\gg\tau_{air}(R)\approx10^{-5}R^2$ can be estimated by setting the asymptotic particle DLC velocities $\Delta V(t)$ equal to the Brownian particle velocities $V_{Br}=(kT/M)^{1/2}$,[9] which are ~10^{-3}-1 m/s for a particle size R~10^{-2}-1 μm, $\rho\approx10^3$ kg/m^3 and $T\approx300$ K. According to Eqs. (5), the viscous air drag force results in asymptotic lift-off distances X_0, corresponding to the product $\Delta V_0\tau_{air}(R)$, with typical size-

dependent X_0 values of $\sim 10^2$ and $\sim 10^4$ μm for $R\sim 1$ μm and $\sim 10^{-2}$ and ~ 1 μm for $R\sim 10^{-2}$ μm at $\Delta V_0 \approx 10$ and $\sim 10^3$ m/s, respectively. These expressions for $X_0 \approx \Delta V_0 \tau_{air}(R)$, $\tau_{air}(R)$ and $D(R)$ were then substituted in equation (4) to yield DLC efficiencies as a function of X_0, R and T_{exp}.

To check the consistency between the model and experiment, $E(\Delta V_0)$ values were calculated for real, non-aggregated ~0.55 and ~0.25 μm polystyrene (PS) particles and $T_{exp} \approx 10^2$ s, corresponding to our single-shot experiments.[3] The resulting curves in Fig.1 exhibit reasonable coincidence with experimental $E(F-F_{DLC})$ values, where F is the fluence of the nanosecond KrF excimer laser.[3] Theoretical DLC efficiencies were then calculated as a function of T_{exp} (Fig.2) for ns and fs/ps laser pulses (typical $\Delta V_0 \sim 10$ m/s and $\sim 10^3$ m/s, respectively) and as a function of ΔV_0 for characteristic exposure times of $\sim 10^{-2}$ and $\sim 10^2$ s (Fig.3). Defining the DLC threshold, F_{DLC}(~10%), at a statistically significant cleaning efficiency value $E \approx 10\%$, DLC of particles, as shown in Fig.2, is possible for 10^{-1}-1 μm particles for ns pulses and for $\sim 10^{-2}$-1 μm particles for ps/fs pulses at characteristic experimental exposure times of $\sim 10^{-2}$-10^2 s and the cleaning efficiency rapidly increases for shorter T_{exp}. DLC of large particles ($R > \sim 0.1$ μm) is possible for ns laser pulses and exposure times of $\sim 10^2$ s, but cleaning of particles smaller than ~0.03 μm requires minimum exposure times $T_{exp} \sim 10^{-2}$ s and maximum lift-off velocities $\Delta V_0 \sim 10^3$ m/s (Fig.3), attainable with ps/fs laser pulses. On the other hand, substrate damage thresholds are lower for ps/fs laser pulses and process windows tend to be narrower.[13] Therefore, for removal of contaminants smaller than $\sim 10^{-2}$ μm from critical surfaces, it may be necessary to employ auxiliary particle trapping tools such as gas jets or suction,[5] and thermophoresis[6]. Steam laser cleaning (SLC)[1,4] provides more efficient particle removal via laser-induced explosive boiling and lift-off of a predeposited liquid layer as the resulting vapor/droplet plumes can produce particle lift-off up to distances of ~ several millimeters.[14]

It should also be noted that in Fig.3 for ~10^{-2}-1 μm particles ΔV_0 values at the 10% level may exceed several times the corresponding critical velocities, V_{crit}, required for particles to leave the near-surface van der Waals potential well of depth, $U(Z)=A\times R/6Z$, e.g., $V_{crit}\approx 1$ m/s$*R^{-1}$ for PS particles on a Si substrate for the Hamaker constant $A_{PS\text{-}Si}\approx 1.2\times 10^{-19}$ J and $Z_{PS\text{-}Si}\approx 0.4$ nm[15] (cf., see Fig.1). Thus, as particle velocities are fluence-dependent for the "trampoline" or "hopping" mechanisms,[1,8] DLC thresholds measured at $E\approx 10\%$ under ambient conditions are actually "effective" thresholds as compared with those, $F_{DLC}(0\%)$, obtained in a low-pressure environment or in vacuum,[2] accounting for both particle-substrate adhesion and particle deceleration in air. For smaller particle sizes, the slopes of measured $E(F)$ curves are small and extrapolation to the actual DLC threshold at $E(F)\approx 0$ is difficult.[2,3]

Furthermore, particle diffusion both to and away from the substrate, starting from the lift-off distance X_0, explains the gradually increasing multi-shot DLC efficiency,[4] resulting in permanent particle depletion in the lift-off space due to outward particle diffusion after many laser irradiation cycles.

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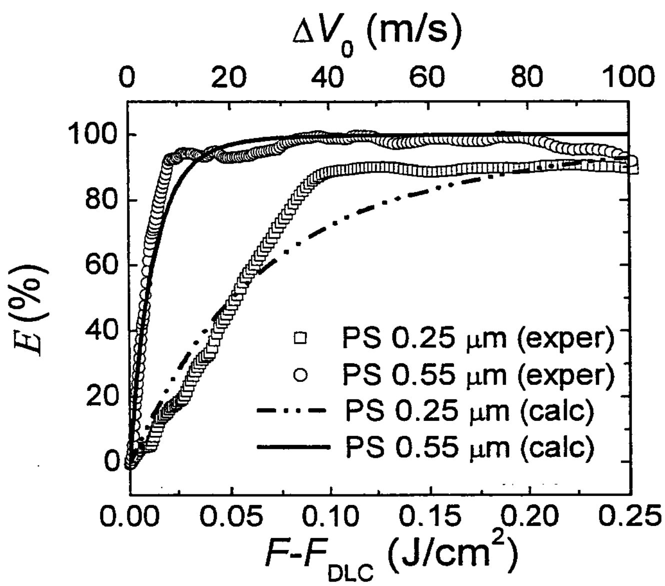
FIG.1. Experimental DLC efficiency, E, as a function of reduced laser fluence, $F-F_{DLC}$, for sub-micron PS particles. DLC thresholds, F_{DLC}, are ~0.02 and ~0.045 J/cm^2 at $E\approx0\%$ for ~0.55 and ~0.25 μm particles, respectively. Calculated E-curves as a function of a lift-off velocity ΔV_0 for an exposure time $T_{exp}\approx10^2$ s.

FIG.2. Calculated DLC efficiency as a function of T_{exp} for ΔV_0 values of ~10 m/s (a) and ~10^3 m/s (b) for different particle sizes. The numbers show particle radii in microns.

FIG.3. Calculated DLC efficiency as a function of ΔV_0 for the characteristic T_{exp} values of ~10^2 s (a) and ~10^{-2} s (b) for different particle sizes. The numbers show particle radii in microns.



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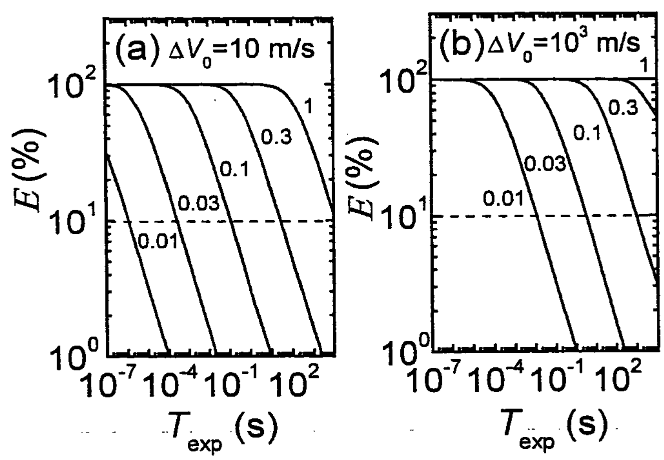

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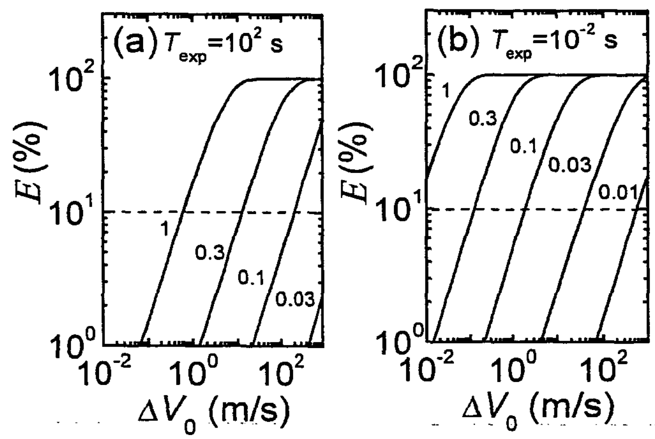

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